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DEFECT CHEMISTRY AND CHARACTERIZATION OF (Hg,Cd)Te

NAS8-33245

Honeywell Electro-Optics Operations, Lexington, Mass.

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I. FORWARD

This semiannual technical report covers the work performed by Honeywell Electro-Optics Operations, Lexington, Massachusetts from January 1, 1981 to June 30, 1981 under the NASA sponsored program entitled "Defect Chemistry and Characterization of (Hg,Cd)Te" on Contract NAS8-33245. The objective of this program is to study and formalize the defect chemistry of (Hg,Cd)Te and to evaluate and select characterization methods for the material.

The principal investigator is Dr. H.R. Vydyanath providing the overall technical direction for the program. Assistance with the experimental work is provided by J.C. Donovan, P. Crickard and A. Barnes. R.A. Lancaster and D.A. Nelson generously furnished all the crystals required for the program.

II. INTRODUCTION

At the end of the first eighteen month period of the program the defect structures of undoped $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ (S), undoped $\text{Hg}_{0.6}\text{Cd}_{0.4}\text{Te}$ (S) copper and indium doped $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ (S) were established. These results were reported in the semiannual technical report covering the period of January 1979 to June 1980. Details of the doping behavior of Iodine doped $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ were reported in the Semiannual technical report covering the period of July 1, 1980 to December 31, 1980. This semiannual report details the work performed from January 1, 1981 to June 30, 1981. During this six month period the defect structure of phosphorus doped $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ has been investigated in detail. Single crystal samples of phosphorus doped $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ were annealed at temperatures varying from 450°C to 600°C in various Hg atmospheres. The samples were quenched to room temperature from the annealing temperatures. Hall effect and mobility measurements were performed at 77K on all these samples. The results indicate the crystals to be p type for a total phosphorus concentration of 10^{19} cm^{-3} in all the samples. The hole concentration at 77K increases with increasing Hg pressures at 450°C and 500°C contrary to the observation in undoped crystals. Also, at low Hg pressures the concentration of holes in the phosphorus doped crystals is lower than in the undoped crystals. The hole concentration in all the samples is lower than the intrinsic carrier concentration at the annealing temperatures. The hole mobility in the doped crystals is similar to that in the undoped crystals. All these inferences have led to the establishment of a defect model according to which phosphorus behaves as a single acceptor interstitially and occupying Te lattice sites while it acts as a single donor occupying Hg lattice sites. Most of the phosphorus appears to be present as neutral $(\text{P}_{\text{Hg}}\text{P}_i)^{\times}$ species at intermediate Hg pressures whereas at low Hg pressures a majority of it is present as $(\text{P}_{\text{Hg}}\text{V}_{\text{Hg}})^{\cdot}$ and $(\text{P}_{\text{Hg}}\text{V}_{\text{Hg}})^{\cdot}$ species. Equilibrium constants have been established for the incorporation of all the phosphorus species. These constants satisfactorily explain all the experimental results.

The following presentations and publications have resulted from the work performed from January 1, 1979 to June 30, 1981.

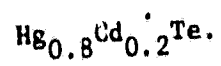
- (1) '(Hg_{1-x}Cd_x)Te Material Studies' H.R. Vydyanath, D.A. Nelson, R.A. Lancaster and R.G. Roy in proceedings of the IRIS Detector Specialty Group meeting, Minneapolis, MN June 15-17, 1979 p. 29.
- (2) 'Defect Chemistry and Characterization of (Hg_{1-x}Cd_x)Te' H.R. Vydyanath, D.A. Nelson and R.A. Lancaster, Journal of Electrochem. Soc. 126, 371C (1979).
- (3) 'Defect Studies in Hg_{0.8}Cd_{0.2}Te' H.R. Vydyanath, presented at the conference on the "Crystal Growth and Characterization of II-VI Compounds", University of Lancaster, U.K. April 14-16, 1980.
- (4) 'Lattice Defects in Hg_{1-x}Cd_xTe Alloys' I-Defect Structure of undoped and copper doped Hg_{0.8}Cd_{0.2}Te, H.R. Vydyanath, J. Electrochem. Soc. 128, 2609 (1981).
- (5) 'Lattice Defects in Hg_{1-x}Cd_xTe Alloys' II-Defect Structure of Indium Doped Hg_{0.8}Cd_{0.2}Te, H.R. Vydyanath, J. Electrochem. Soc. 128, 2619 (1981).
- (6) 'Lattice Defects in Hg_{1-x}Cd_xTe Alloys' III-Defect Structure of Undoped Hg_{0.6}Cd_{0.4}Te, H.R. Vydyanath, J.C. Donovan and D.A. Nelson, J. Electrochem Soc. 128, 2625 (1981).
- (7) 'Doping behavior of Iodine in Hg_{0.8}Cd_{0.2}Te' H.R. Vydyanath and F.A. Kroger, J. Electronic Materials Vol. 11, No. 1 (January 1982 issue)
- (8) 'Mode of Incorporation of Phosphorus in Hg_{0.8}Cd_{0.2}Te' H.R. Vydyanath and R.C. Abbott, J. Appl. Phys. (submitted November 1981).
- (9) 'Role of lattice Defects in Undoped and Doped Hg_{1-x}Cd_xTe Alloys' H.R. Vydyanath HCT Workshop Minneapolis, October 28-30, 1981.

III. TECHNICAL DETAILS

The work performed from January 1, 1981 to June 30, 1981 resulted in the establishment of a defect model for phosphorus doped $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$. The technical details of the work are described in the form of a paper entitled 'Mode of Incorporation of Phosphorus in $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ ' which is being submitted to the Journal of Applied Physics for publication. The paper describes the background material, the experimental details and analysis of the data. The mass action constants arrived at for the incorporation of phosphorus in $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ together with the values of the constants for the intrinsic excitation process and the incorporation of the doubly ionized native acceptor defect (established in one previous semiannual report) satisfactorily explain the experimental results for phosphorus doped $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ crystals.

III.1

DEFECT CHEMISTRY AND CHARACTERIZATION OF PHOSPHORUS DOPED



Mode of Incorporation of Phosphorus in $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ *

H.R. Vydyanath and R.C. Abbott

Honeywell, Inc.

Electro Optics Operations

2 Forbes Road

Lexington, Mass. 02173

Single crystal samples of phosphorus doped $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ were annealed at temperatures varying from 450°C to 600°C in different partial pressures of Hg. Hall effect and mobility measurements were performed on the samples cooled to room temperature. All the samples were found to be p-type with the hole concentration being much less than the total amount of phosphorus present in the crystals. The hole concentration was found to increase with increase in partial pressure of Hg in contrast to the behavior observed in undoped crystals. Also, the hole concentration obtained in the doped samples at low Hg pressures was less than that in undoped crystals. The 77 K hole mobility of the doped samples was similar to that of undoped samples. All these inferences indicate that phosphorus behaves amphoterically in $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te(s)}$ acting as a single acceptor occupying interstitial and tellurium lattice sites at high Hg pressures and as a single donor occupying Hg lattice sites at low Hg pressures; at intermediate Hg pressures, the majority of the phosphorus appears to be present as electrically neutral pairs formed from the association of the interstitial and substitutional phosphorus species, $(\text{P}_i\text{PHg})^X$. At low Hg pressures, a large fraction of the phosphorus appears to be present as $(\text{PHgVHg})'$ and $(\text{PHgVHg})^\bullet$.

Thermodynamic constants evaluated for the incorporation of the various phosphorus species satisfactorily explain the experimental results.

Key words: II-VI compounds, $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$, phosphorus doping, hole mobility, defects, ionized impurity scattering.

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I. INTRODUCTION

The mode of incorporation of phosphorus in CdTe has been studied in detail by Selim and Kröger [1] who correlated their own electrical data with the diffusion and solubility data of Hall and Woodbury [2] to arrive at the defect state. According to their findings, phosphorus behaves amphoterically in CdTe acting as an acceptor interstitially (P_i^{\cdot}) and on Te lattice sites (P_{Te}^{\cdot}) and as a triple donor on Cd lattice sites ($PCd^{\cdot\cdot\cdot}$); at high phosphorus concentrations, a large fraction was found to be present as neutral associates ($PCdP_i$)^x. In this paper we have undertaken to examine the role of phosphorus in $Hg_{0.8}Cd_{0.2}Te$ via Hall effect and mobility measurements on phosphorus doped crystals quenched from 450-600°C subsequent to anneals in different partial pressures of Hg. The results indicate that the behavior of phosphorus in $Hg_{0.8}Cd_{0.2}Te$ is similar to that established for CdTe [1] except that all the electrically active phosphorus defect centers in $Hg_{0.8}Cd_{0.2}Te$ appear to be only singly ionized. At low Hg pressure, phosphorus is incorporated as a single donor occupying Hg lattice sites (P_{Hg}^{\cdot}) and at high Hg pressure as a single acceptor on interstitial sites (P_i^{\cdot}) and Te lattice sites (P_{Te}^{\cdot}). At moderate Hg pressures, a large fraction is found to be present as neutral pairs ($P_{Hg}P_i$)^x. At low Hg pressures, electrically active associates ($P_{Hg}V_{Hg}$)' and ($P_{Hg}V_{Hg}$)[•] appear to be present in appreciable concentrations. The equilibrium constants established for the incorporation of the various phosphorus defects explain the experimental results satisfactorily.

II. EXPERIMENTAL

A. Preparation of the Phosphorus Doped Crystals

A phosphorus doped $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ ingot was grown by the solid state recrystallization method [3]. Phosphorus corresponding to a concentration of 10^{19}cm^{-3} was added to the starting charge. Single crystal slices were then cut from the boule. The slices were lapped, polished, etched in Br methanol and rinsed in DI water prior to the anneals.

B. Hg Vapor Anneals

The samples were annealed in evacuated quartz ampoules containing some Hg to obtain the desired Hg pressure [4]. In isothermal anneals, the partial pressure of Hg was dependent on the amount of Hg and the volume of the ampoule. In non-isothermal anneals, the partial pressure of Hg was determined from the temperature of the Hg. The limits of Hg pressure-within which $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te(s)}$ is stable--were obtained from the partial pressure data of Tung et al. [5]. In order to assure ourselves of equilibration within reasonable annealing times, the sample thicknesses were restricted to less than 0.04 cm. Annealing times ranged from 72 hours at 550 to 600°C to 7 days at 500°C and approximately 4 to 6 weeks at 450°C. These annealing times appeared to be satisfactory for complete equilibration. Subsequent to the equilibration in Hg vapor, the ampoule containing the sample was quenched in ice water.

C. Electrical Measurements

Hall effect and electrical resistivity measurements were carried out using the Van der Pauw method [6]. Magnetic field strengths of 400 gauss and 4000 gauss were used for the measurement of the Hall coefficient.

D. Chemical Analysis

The concentration of phosphorus in the samples was mass spectrographically analyzed (Photometrics Inc., Woburn, Mass.) and the concentration determined from the analysis was close to what had been added to the starting charge within $\pm 20\%$.

III. Results

Only those Hall effect data where the Hall coefficient did not vary with the magnetic field were used in evaluating the carrier concentration. This procedure ensured that the samples did not exhibit mixed conduction [7] and thus an unambiguous evaluation of the carrier concentration was possible. The carrier concentration was evaluated using the expression:

$$n \text{ or } p = \frac{1}{|R_H|q}$$

Figure 1 shows the Hall coefficient as a function of the temperature of measurement from temperatures below 77 K to 300 K for phosphorus doped samples annealed at various temperatures in different partial pressure of Hg. The data indicate that the phosphorus centers are all ionized at 77 K;

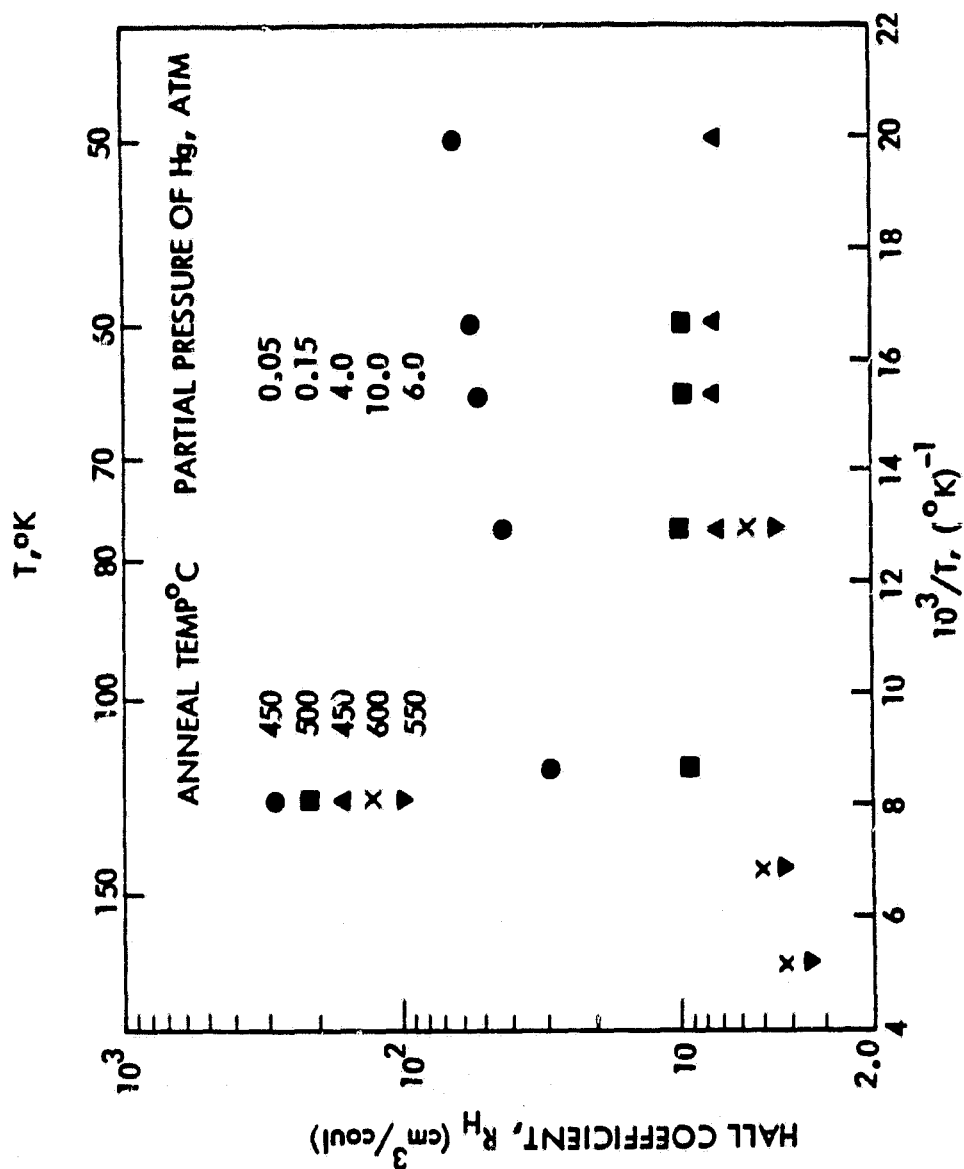


Figure 1. Hall Coefficient as a Function of Temperature of Measurement for Phosphorus Doped (10^{19} cm^{-3}) $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ Crystals Annealed at Different Temperatures under Various Partial Pressures of Hg and Quenched to Room Temperature.

therefore all the Hall measurements were carried out at 77 K and the resulting hole concentration was assumed to give a measure of the concentration of the defects at the higher annealing temperatures.

Figure 2 shows the hole concentrations obtained in samples doped with 10^{19}cm^{-3} of phosphorus which were annealed at various temperatures in different partial pressures of Hg; the data shown in Figure 2 are replotted in Figures 3 through 6 for different temperatures of anneal. Additional data shown in these figures include the hole concentration and hole mobility obtained in the undoped samples as well as the hole mobility in the phosphorus doped samples, all of which are plotted as a function of the partial pressure of Hg.

Several inferences can be made from the results of Figures 2 through 6. They are:

- (1) The hole concentration in the phosphorus doped samples is lower than the total concentration of phosphorus in the samples and lower than the intrinsic carrier concentration for $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te(s)}$ at the temperatures of annealing reported here [4].
- (2) The hole mobility in the phosphorus doped samples increases with increasing partial pressure of Hg just as is the case for the undoped samples.

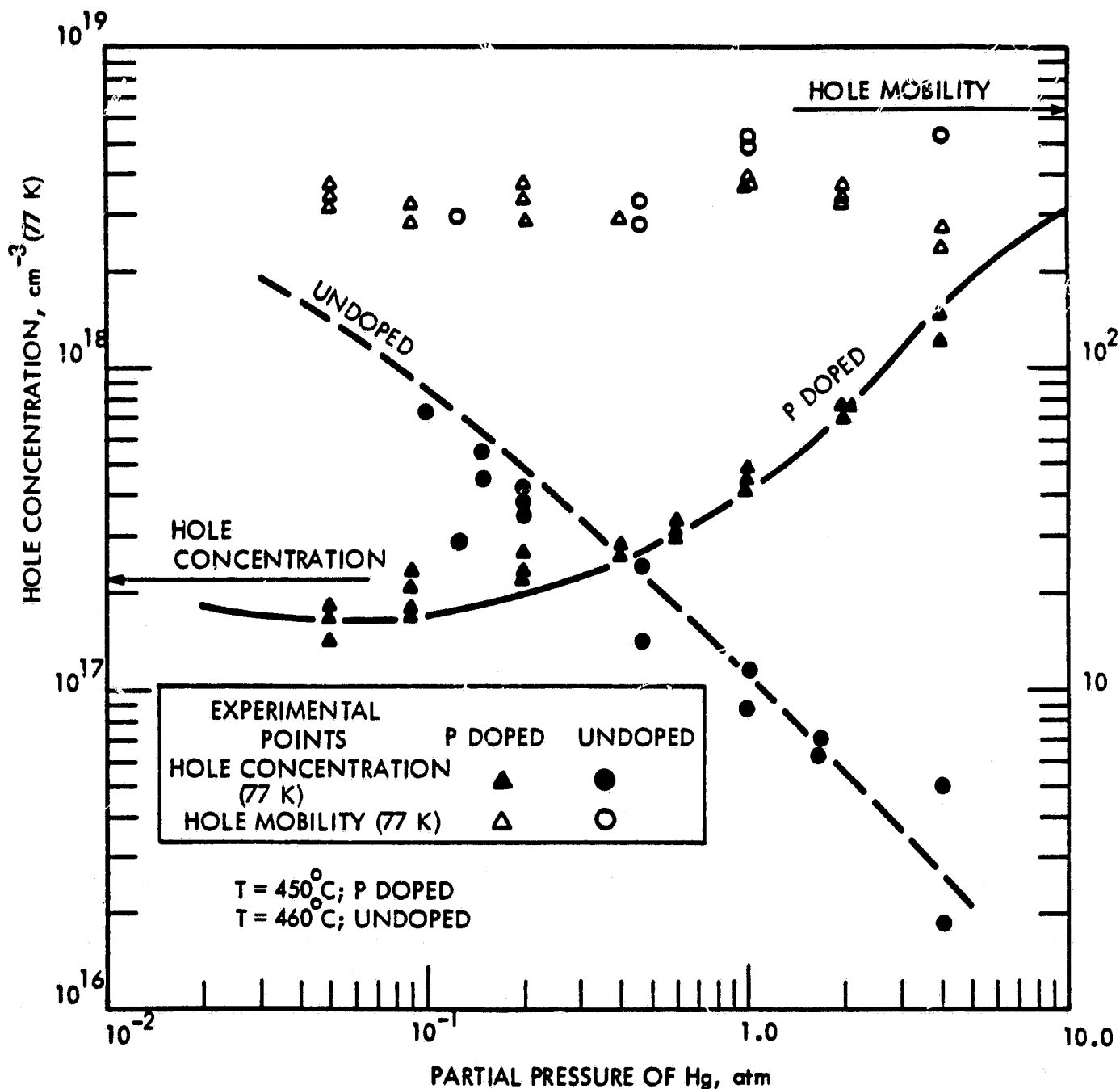


Figure 3. Hole Concentration and Hole Mobility at 77 K as a Function of the Partial Pressure of Hg for Phosphorus Doped (10^{19}cm^{-3}) $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ Crystals Quenched to Room Temperature After Annealing 450°C . Similar data are shown for undoped samples annealed at 460°C [Reference 4]. The solid and the dashed lines show the calculated hole concentrations to be expected in the doped and the undoped samples on the basis of the defect model.

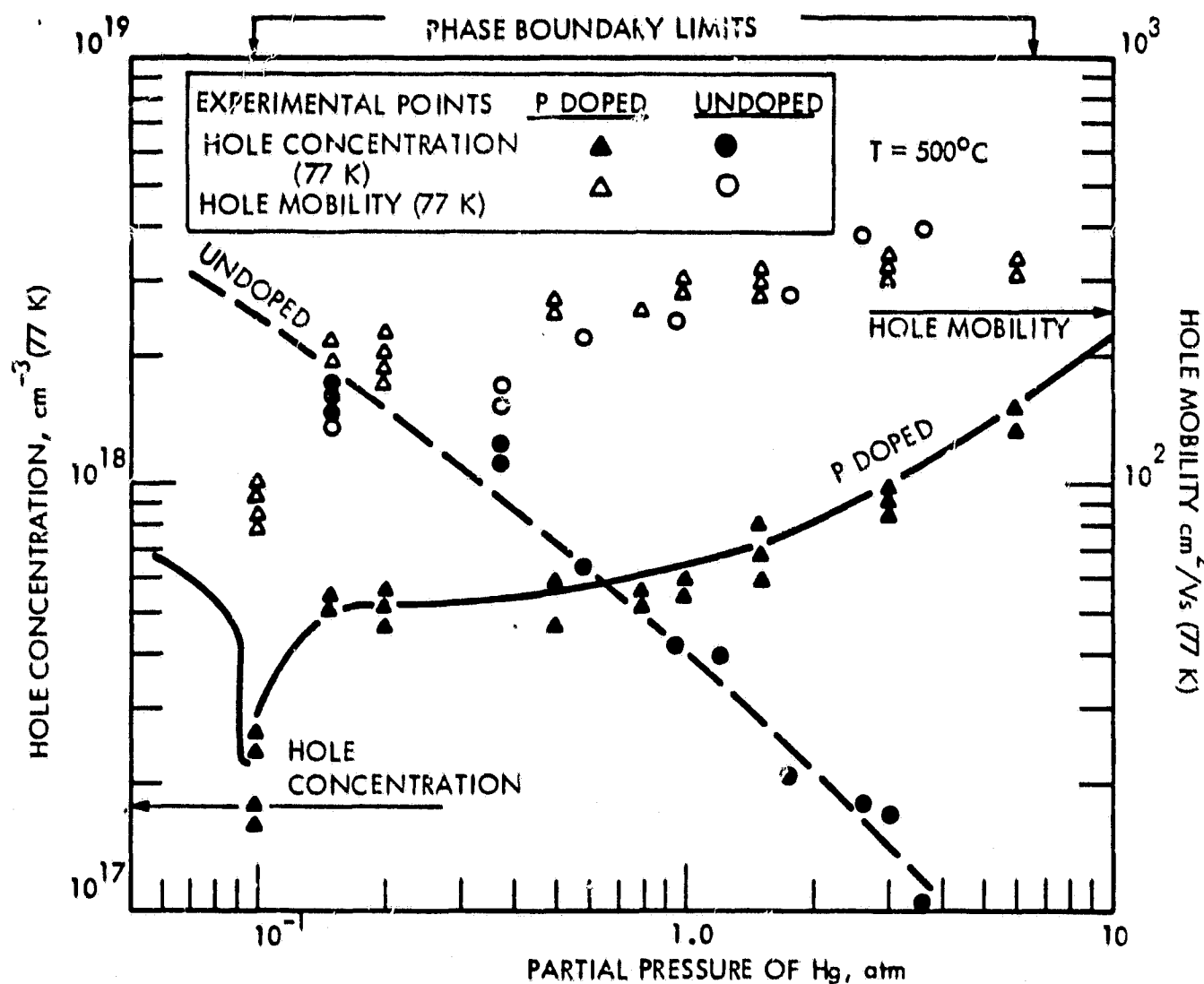


Figure 4. Hole Concentration and Hole Mobility at 77 K as a Function of the Partial Pressure of Hg for Phosphorus Doped (10^{19}cm^{-3}) and Undoped [Reference 4] $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ Crystals Quenched to Room Temperature After Equilibration at 500°C ; the solid and the dashed lines show the calculated hole concentrations to be expected in the doped and the undoped samples on the basis of the defect model.

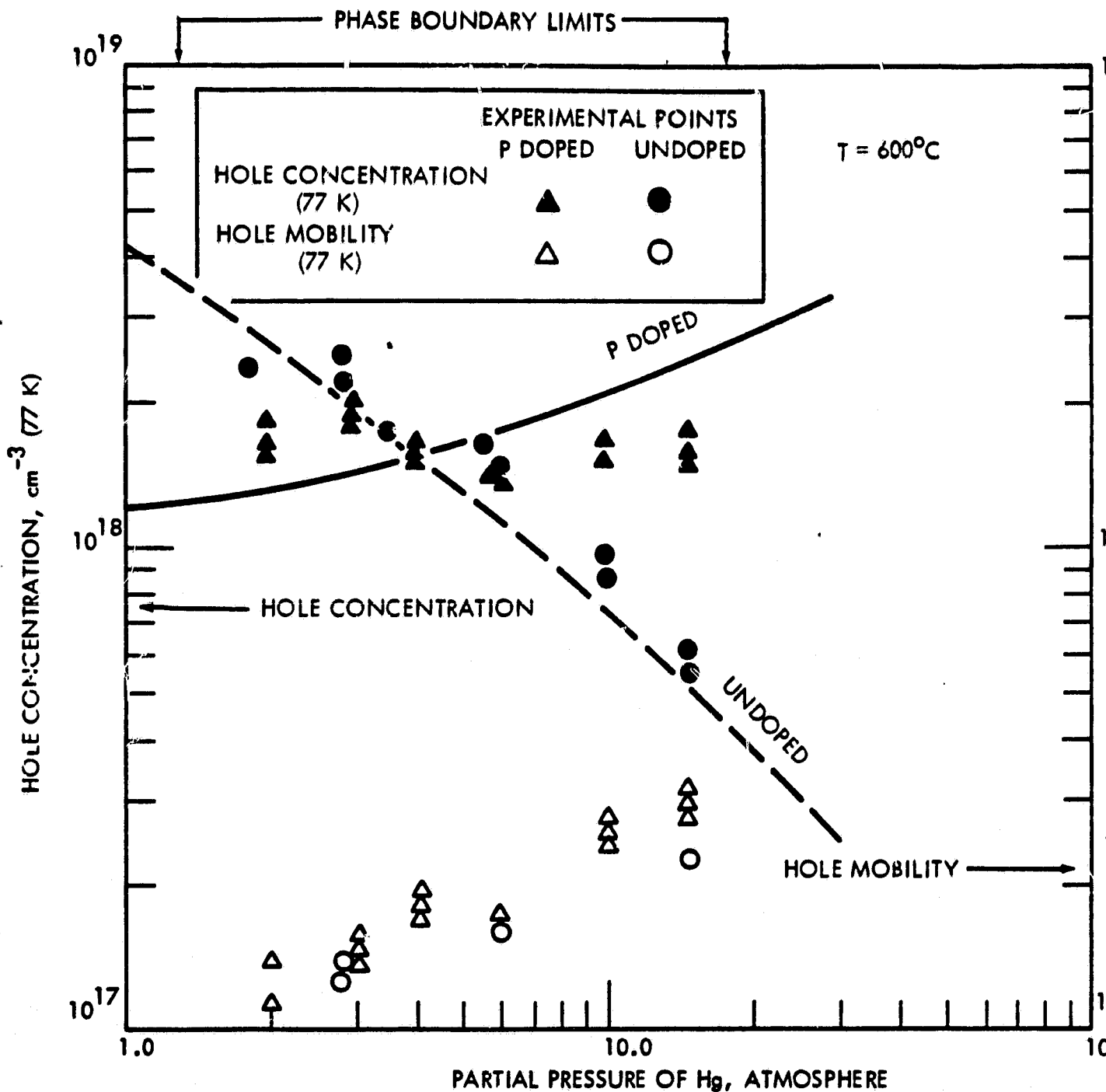


Figure 6. Hole Concentration and Hole Mobility at 77 K as a Function of the Partial Pressure of hg for Phosphorus Doped (10^{19}cm^{-3}) and Undoped [Reference 4] $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ Crystals Quenched to Room Temperature After Equilibration at 600°C ; the solid and the dashed lines show the calculated hole concentrations to be expected in the doped and the undoped samples on the basis of the defect model.

- (3) The hole concentration in the phosphorus doped samples increases with increasing partial pressure of Hg; the hole concentrations at high Hg pressures are higher than found in the undoped crystals.
- (4) The hole concentration in the samples annealed at 500°C and 450°C under very low Hg pressures is less than that obtained in the undoped samples for comparable annealing conditions.

equation are all listed in Table I. Only those defects which were found to be important as a result of the present work are shown in Table I. Table II lists the exponents of the partial pressure of Hg and the phosphorus concentration for all the defects for various approximations to the electroneutrality condition and the phosphorus balance equation.

Table I. DEFECT FORMATION REACTIONS, MASS ACTION RELATIONS, ELECTRONEUTRALITY CONDITION AND PHOSPHOROUS BALANCE EQUATION

Reaction	Mass Action Relation
1. $0 \rightarrow e^- + h^+$	$K_i = [e^-][h^+]$
2. $Hg^X \rightarrow V_{Hg}^{..} + 2h^+ + Hg(g)$	$K_{V_{Hg}}^{..} = [V_{Hg}^{..}][h^+]^2 p_{Hg}$
3. $2h^+ + (P_i P_{Hg})^X + Hg_{Hg}^X \rightarrow 2 P_{Hg}^{\cdot} + Hg(g)$	$K_{P_{Hg}}^{\cdot} = [P_{Hg}^{\cdot}]^2 p_{Hg} / [h^+]^2 [(P_i P_{Hg})^X]$
4. $(P_i P_{Hg})^X + 3 Hg(g) \rightarrow 3 Hg_{Hg}^X + 2 P_{Te}^{\cdot} + 2 h^+$	$K_{P_{Te}}^{\cdot} = [P_{Te}^{\cdot}]^2 [h^+]^2 / [(P_i P_{Hg})^X]^3 p_{Hg}$
5. $P_i^{\cdot} + P_{Hg}^{\cdot} \rightarrow (P_i P_{Hg})^X$	$K_P (P_i P_{Hg})^X = [(P_i P_{Hg})^X] / [P_i^{\cdot}] [P_{Hg}^{\cdot}]$
6. $P_{Hg}^{\cdot} + V_{Hg}^{..} \rightarrow (P_{Hg} V_{Hg})^{\cdot}$	$K_P (P_{Hg} V_{Hg})^{\cdot} = [(P_{Hg} V_{Hg})^{\cdot}] / [P_{Hg}^{\cdot}] [V_{Hg}^{..}]$
7. $(P_i P_{Hg})^X + 2h^+ + 3 Hg_{Hg}^X \rightarrow 2(P_{Hg} V_{Hg})^{\cdot} + 3 Hg(g)$	$K (P_{Hg} V_{Hg})^{\cdot} = [(P_{Hg} V_{Hg})^{\cdot}]^2 p_{Hg}^3 / [(P_i P_{Hg})^X] [h^+]^2$

Electroneutrality Condition

$$[e^-] + 2 [V_{Hg}^{..}] + [P_i^{\cdot}] + [P_{Te}^{\cdot}] + [(P_{Hg} V_{Hg})^{\cdot}] = [h^+] + [P_{Hg}^{\cdot}] + [(P_{Hg} V_{Hg})^{\cdot}]$$

Phosphorous Balance Equation

$$[P_i^{\cdot}] + [P_{Hg}^{\cdot}] + [P_{Te}^{\cdot}] + [(P_{Hg} V_{Hg})^{\cdot}] + [(P_{Hg} V_{Hg})^{\cdot}] + 2 [(P_i P_{Hg})^X] = [P_{Tot}]$$

Table II. VARIATIONS OF THE DEFECT CONCENTRATIONS AS A FUNCTION OF p_{Hg} AND/OR PHOSPHORUS CONCENTRATIONS FOR VARIOUS APPROXIMATIONS TO THE ELECTRONEUTRALITY CONDITION AND PHOSPHORUS BALANCE EQUATION

	Type of defect and approximation to the electroneutrality condition and phosphorus balance equation	$[]_{\alpha} p_{\text{Hg}} [p_{\text{Tot}}]^s$		
		r	r	s
(1)	$[e^{\cdot}] = [h^{\cdot}] = K_i$			
	$[(P_i P_{\text{Hg}})^{\times}] = [P_{\text{Tot}}]/2$	0		1
	$[V_{\text{Hg}}^{\cdot\cdot}]$	-1		0
	$[P_i^{\cdot}]$	1/2		1/2
	$[P_{\text{Hg}}^{\cdot}]$	-1/2		1/2
	$[P_{\text{Te}}^{\cdot}]$	3/2		1/2
	$[(P_{\text{Hg}} V_{\text{Hg}})^{\cdot}]$	-3/2		1/2
	$[(P_{\text{Hg}} V_{\text{Hg}})^{\cdot\cdot}]$	-3/2		1/2

B. Choice of a Defect Model

Based on the fact that P belongs to group V, Hg to group II and Te to group VI, a number of different phosphorus species can be expected depending on the type of lattice sites occupied. These are:

Interstitially $P_i^{\cdot}, P_i^{\cdot\cdot} \dots P_i^{\cdot\cdot\cdot\cdot}$ (Acceptors)

Substitutionally $P_{Hg}^{\cdot}, P_{Hg}^{\cdot\cdot}$ and $P_{Hg}^{\cdot\cdot\cdot}$ (Donors)
(occupying Hg lattice site)

Substitutionally P_{Te}^{\cdot} (Acceptors)
(occupying Te lattice site)

Pairs - a) $(P_i P_{Hg})^X, (P_i P_{Hg})^{\cdot}, (P_i P_{Hg})^{\cdot\cdot}$
 $(P_i P_{Hg})^{\cdot\cdot\cdot}, (P_i P_{Hg})^{\cdot\cdot\cdot\cdot}, (P_i P_{Hg})^{\cdot\cdot\cdot\cdot\cdot}, (P_i P_{Hg})^{\cdot\cdot\cdot\cdot\cdot\cdot}$

b) $(P_{Hg} V_{Hg})^X, (P_{Hg} V_{Hg})^{\cdot}$ and $(P_{Hg} V_{Hg})^{\cdot\cdot}$

c) $(P_{Hg} P_{Te})^X, (P_{Hg} P_{Te})^{\cdot}$ and $(P_{Hg} P_{Te})^{\cdot\cdot}$

The data shown in Figures 3 through 6 indicate that the hole mobility values obtained in the phosphorus doped samples are comparable to those in the undoped samples for similar partial pressures of Hg and temperatures of annealing even though at high Hg pressures, the hole concentrations in the phosphorus doped samples are higher than in the undoped samples. This inference already precludes the possibility of the presence of multiply electrically charged phosphorus species in large concentrations since such species can be expected to behave as much stronger scattering centers than the singly charged species,

thus causing the hole mobility in the phosphorus doped samples to be considerably lower than the hole mobility in the undoped samples contrary to the experimental results. The species of interest are reduced to

$P_i^{\cdot}, P_{Hg}^{\cdot}, P_{Te}^{\cdot}, (P_{Hg}V_{Hg})^{\cdot}, (P_{Hg}V_{Hg})^{\cdot}, (P_{Hg}P_{Te})^{\cdot}, (P_iP_{Hg})^X, (P_{Hg}V_{Hg})^X$ and $(P_{Hg}P_{Te})^X$. The fact that the hole concentrations obtained in the phosphorus doped samples are much lower than the total phosphorus concentration in the samples (10^{19} cm^{-3}) indicates that most of the phosphorus is present as neutral species in the form of $(P_iP_{Hg})^X$, $(P_{Hg}V_{Hg})^X$ or $(P_{Hg}P_{Te})^X$. It is also to be noted that the hole concentrations obtained in the phosphorus doped samples (Figures 2 through 6) are smaller than the intrinsic carrier concentrations to be expected at the temperatures of anneal reported here [4].

The choice of a defect model is reduced to:

$$(1) [e'] = [h^{\cdot}] = \sqrt{K_i} \neq f(p_{Hg}) \text{ and } [(P_{Hg}P_{Te})^X] = [P_{Tot}/2]$$

$$(2) [e'] = [h^{\cdot}] = \sqrt{K_i} \neq f(p_{Hg}) \text{ and } [(P_{Hg}V_{Hg})^X] = [P_{Tot}]$$

and $(3) [e'] = [h^{\cdot}] = \sqrt{K_i} \neq f(p_{Hg}) \text{ and } [(P_iP_{Hg})^X] = [P_{Tot}]/2$

The expected power dependences of the various defects on the partial pressure of Hg for the above defect model situations are listed in Table II. Models 1 and 2 for which $[P_{Tot}/2] = [(P_{Hg}P_{Te})^X]$ or $[(P_{Hg}V_{Hg})^X]/2$ predict much stronger dependences of $[P_{Hg}^{\cdot}]$ on p_{Hg} and the search did not result in a consistent set of mass action constants for the incorporation of the various phosphorus species which could explain the experimental results of Figures 3

through 6. The third defect model for which $[e'] = [h'] = \sqrt{K_i} * f(P_{Hg})$ and $[(P_i P_{Hg})^x] = [P_{Tot}]/2$ successfully explains the experimental results of Figures 3 through 6; for such a situation it can be deduced from the mass action relations of Table I that

$$[P_i'] \propto P_{Hg}^{1/2}, [P_{Hg}^\bullet] \propto P_{Hg}^{-1/2}, [P_{Te}'] \propto P_{Hg}^{3/2}, [(P_{Hg} V_{Hg})'] \propto P_{Hg}^{-3/2}$$

and $[(P_{Hg} V_{Hg})^\bullet] \propto P_{Hg}^{-3/2}$. These exponents are listed in Table II. It should be mentioned here that although our initial considerations for the electrically active phosphorus species were restricted to the simple species, P_i' , P_{Hg}^\bullet and P_{Te}' , it was immediately realized that the experimental results warranted the introduction of the species $(P_{Hg} V_{Hg})'$ and $(P_{Hg} V_{Hg})^\bullet$ also.

The pairs $(P_{Hg} V_{Hg})^\bullet$ are formed from the association of the species $P_{Hg}^{\bullet\bullet}$ and V_{Hg}' whereas P_{Hg}^\bullet and V_{Hg}'' make up the pairs $(P_{Hg} V_{Hg})'$.

The explanation for the presence of appreciable concentrations of $(P_{Hg} V_{Hg})^\bullet$ without the presence of large concentrations of isolated $P_{Hg}^{\bullet\bullet}$ species lies in the fact that the donor energy level of the $P_{Hg}^{\bullet\bullet}$ species (which upon ionization gives $P_{Hg}^{\bullet\bullet}$) may lie outside the forbidden gap (in the valence band) and pairing may bring its level to within the gap [13]. Examples of such level variations upon pairing have been found by us earlier for Cds: Ag [14] and Si: Se [15].

C. DEFECT MODEL CALCULATIONS AND COMPARISON WITH EXPERIMENTAL RESULTS

In this section we examine the validity of the proposed defect model for phosphorus doped $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ from a comparison of the electrical data with those one calculates from the defect model. Although the defect model arrived at in the previous section took into consideration the mobility data also, it will become apparent from discussions in this section that the agreement between the experimental values and calculations (based on the defect model) is better for carrier concentration than for hole mobilities.

C.1 Analysis of the Carrier Concentration

According to the defect model deduced in the previous section, the phosphorus species of interest are P_i^+ , P_{Hg}^\bullet , P_{Te}^+ , $(P_{\text{Hg}}V_{\text{Hg}})'$, $(P_{\text{Hg}}V_{\text{Hg}})^\bullet$ and $(P_iP_{\text{Hg}})^x$.

The complete electroneutrality condition is written as:

$$[e^-] + 2 [V_{\text{Hg}}''] + [P_i^+] + [P_{\text{Te}}^+] + [(P_{\text{Hg}}V_{\text{Hg}})'] = [h^\bullet] + [P_{\text{Hg}}^\bullet] + [(P_{\text{Hg}}V_{\text{Hg}})^\bullet] \quad (1)$$

Expressing all the species in terms of $[h^\bullet]$, p_{Hg} and $[(P_iP_{\text{Hg}})^x]$ via the mass action relations listed in Table 1, we get:

$$\begin{aligned} & \frac{K_1}{[h^\bullet]} + \frac{2 K_{V_{\text{Hg}}}''}{p_{\text{Hg}} [h^\bullet]^2} + \frac{[(P_iP_{\text{Hg}})^x]^{1/2} p_{\text{Hg}}^{1/2}}{K_p (P_iP_{\text{Hg}})^x K_{P_{\text{Hg}}}^{1/2} [h^\bullet]} \\ & + \frac{K_{P_{\text{Te}}}^{1/2} p_{\text{Hg}}^{3/2} [(P_iP_{\text{Hg}})^x]^{1/2}}{[h^\bullet]} \end{aligned}$$

$$\begin{aligned}
& + \frac{K_P (P_{Hg} V_{Hg})', K_{P_{Hg}} [(P_i P_{Hg})^X]^{1/2} K_{V_{Hg}}''}{[h^\bullet] P_{Hg}^{3/2}} \\
& - [h^\bullet] - \frac{K_{P_{Hg}} [h^\bullet] [(P_i P_{Hg})^X]^{1/2}}{P_{Hg}^{1/2}} \\
& - \frac{K_{(P_{Hg} V_{Hg})}^{1/2} \cdot [h^\bullet] [(P_i P_{Hg})^X]^{1/2}}{P_{Hg}^{3/2}} = 0
\end{aligned}
\quad \text{-- (2)}$$

The phosphorus balance equation is written as:

$$\begin{aligned}
& [P_i'] + [P_{Hg}^\bullet] + [P_{Te}'] + [(P_{Hg} V_{Hg})'] \\
& + [(P_{Hg} V_{Hg})^\bullet] + 2 [(P_i P_{Hg})^X] = [P_{Tot}]
\end{aligned}
\quad \text{-- (3)}$$

The concentrations of the species P_i' , P_{Te}' , $(P_{Hg} V_{Hg})'$, P_{Hg}^\bullet and $(P_{Hg} V_{Hg})^\bullet$ are respectively given by the III, IV, V, VII and VIII terms of equation (2).

The concentration of holes obtained in the crystals cooled to 77 K is given by:

$$\begin{aligned}
P(77 \text{ K}) &= [P_i'] + [P_{Te}'] + [(P_{Hg} V_{Hg})'] + 2 [V_{Hg}'] \\
&\quad - [P_{Hg}^\bullet] - [(P_{Hg} V_{Hg})^\bullet]
\end{aligned}
\quad \text{-- (4)}$$

(The assumption made here is that the electrons and holes recombine during quenching and only the atomic defects are frozen in [8-9].)

The concentrations $[P_i']$, $[P_{Te}']$, $[(P_{Hg}V_{Hg})']$, $2[V_{Hg}^{II}]$, $[P_{Hg}^{\bullet}]$ and $[(P_{Hg}V_{Hg})^{\bullet}]$ in equation 4 are respectively given by the III, IV, V, II, VII and VIII terms of equation (2).

Of the mass action constants K_i , $K_{V_{Hg}}^{II}$, $K_{P_{Hg}}$, $K_{P_{Te}}$, $K_{P(P_{Hg}V_{Hg})'}$ and $K_{(P_{Hg}V_{Hg})^{\bullet}}$, K_i and $K_{V_{Hg}}^{II}$ are known from earlier work on the undoped crystals [4] and are given by:

$$K_i = 9.16 \times 10^{40} \exp (-0.57 \text{ eV}/K_T) \text{ cm}^{-6} \quad \text{-- (5)}$$

and

$$K_{V_{Hg}}^{II} = 1.58 \times 10^{69} \exp (-2.24 \text{ eV}/K_T) \text{ cm}^{-9} \text{ atm} \quad \text{-- (6)}$$

With the knowledge of the constants K_i and $K_{V_{Hg}}^{II}$ and $[P_{Tot}] = 10^{19} \text{ cm}^{-3}$, a procedure of trial and error was used to arrive at the values of the other mass action constants such that a solution to equations 2 and 3 yielded hole concentrations at 77 K (using equation 4) which agreed best with the experimentally observed values; the concentrations of all the defects present in the crystal are also immediately obtained from the various terms in equation (2). The hole concentrations calculated from the defect model are shown in Figures 3 through 6 as solid lines; the agreement between the calculations and the experimental values appears satisfactory particularly at 450 C and 500 C. The agreement is not as good at 550 C and 600 C probably due to quenching inefficiency [4] at the higher temperatures. The calculated defect isotherms for various annealing temperatures are shown in Figures 7 through 10. The

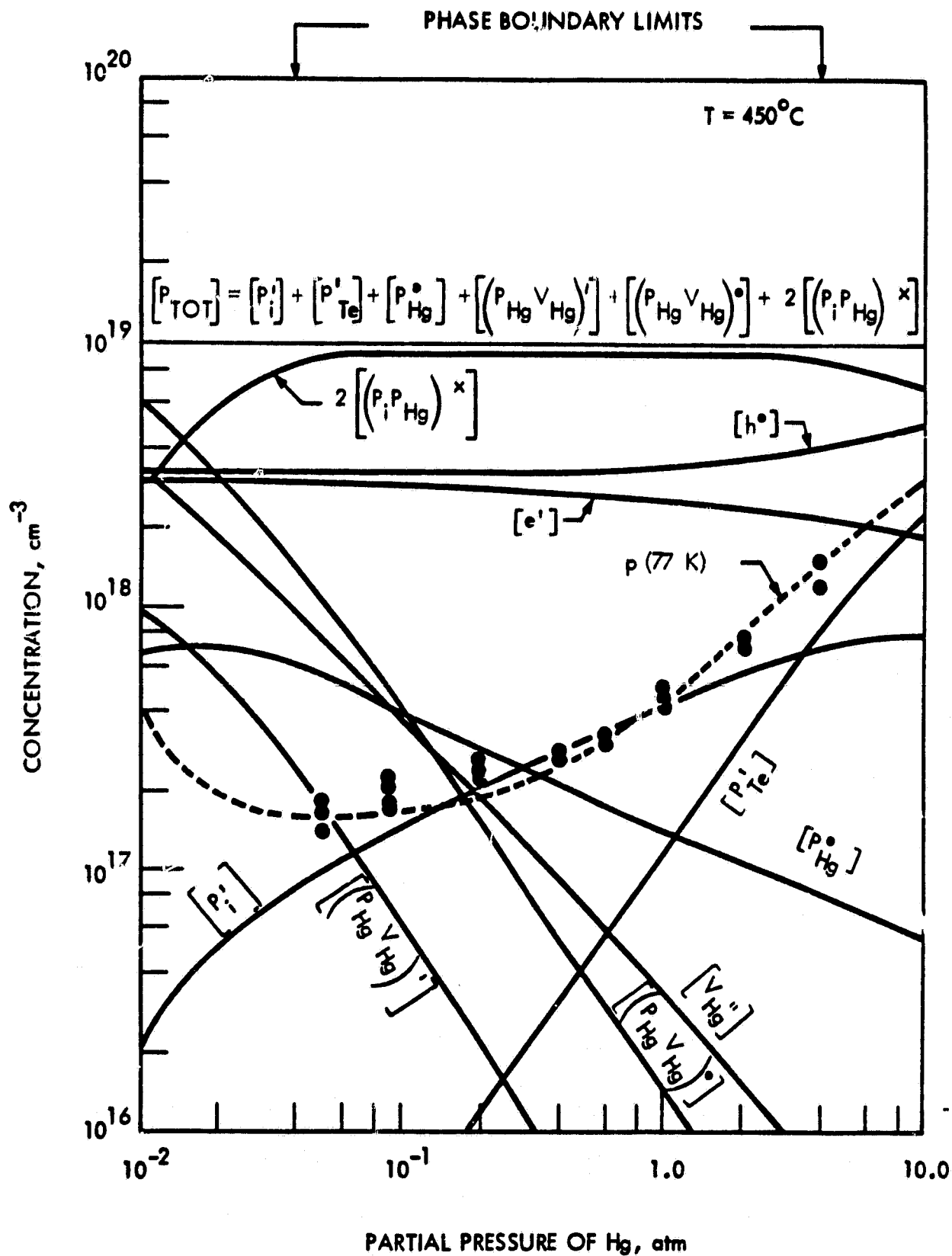


Figure 7. Calculated Concentrations of $[e']$, $[h^\bullet]$, $[V_{\text{Hg}}^{\text{II}}]$, $[P_i']$, $[P_{\text{Te}}']$, $[P_{\text{Hg}}^\bullet]$, $[(P_{\text{Hg}}V_{\text{Hg}})^\bullet]$, $[(P_{\text{Hg}}V_{\text{Hg}})']$ and $[(P_iP_{\text{Hg}})^\times]$ as a Function of the Partial Pressure of Hg at 450°C ; the expected hole concentration in the crystals cooled to 77 K is indicated as a dashed line along with the experimental results.

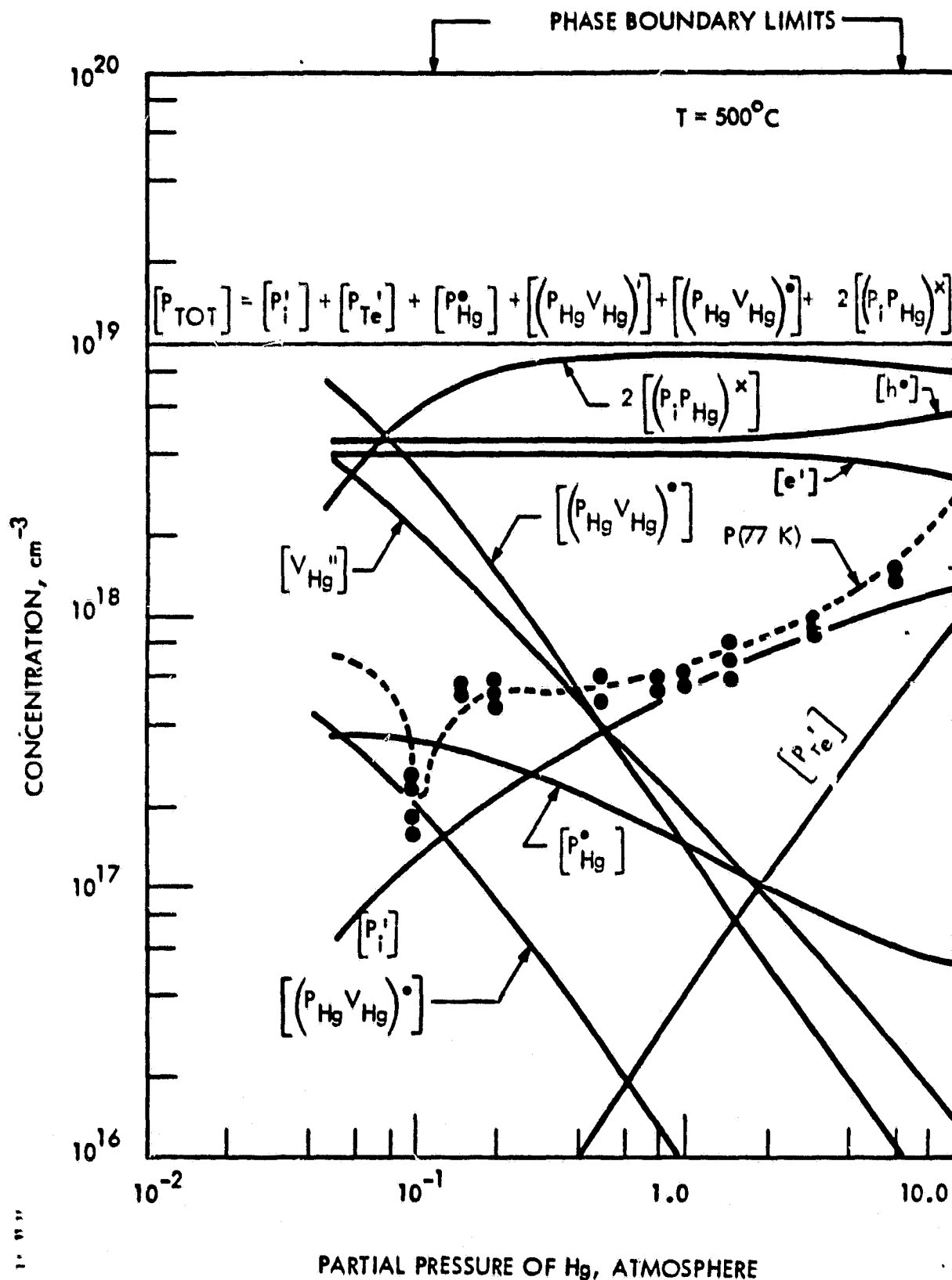


Figure 8. Calculated Concentrations of $[e']$, $[h^\bullet]$, $[V_{\text{Hg}}^{\text{W}}]$, $[P_i']$, $[P_{\text{Te}}']$, $[P_{\text{Hg}}^\bullet]$, $[(P_{\text{Hg}}V_{\text{Hg}})^\bullet]$, $[(P_{\text{Hg}}V_{\text{Hg}})']$ and $[(P_iP_{\text{Hg}})^x]$ as a Function of the Partial Pressure of Hg at 500°C ; the expected hole concentration in the crystals cooled to 77 K is indicated as a dashed line along with the experimental results.

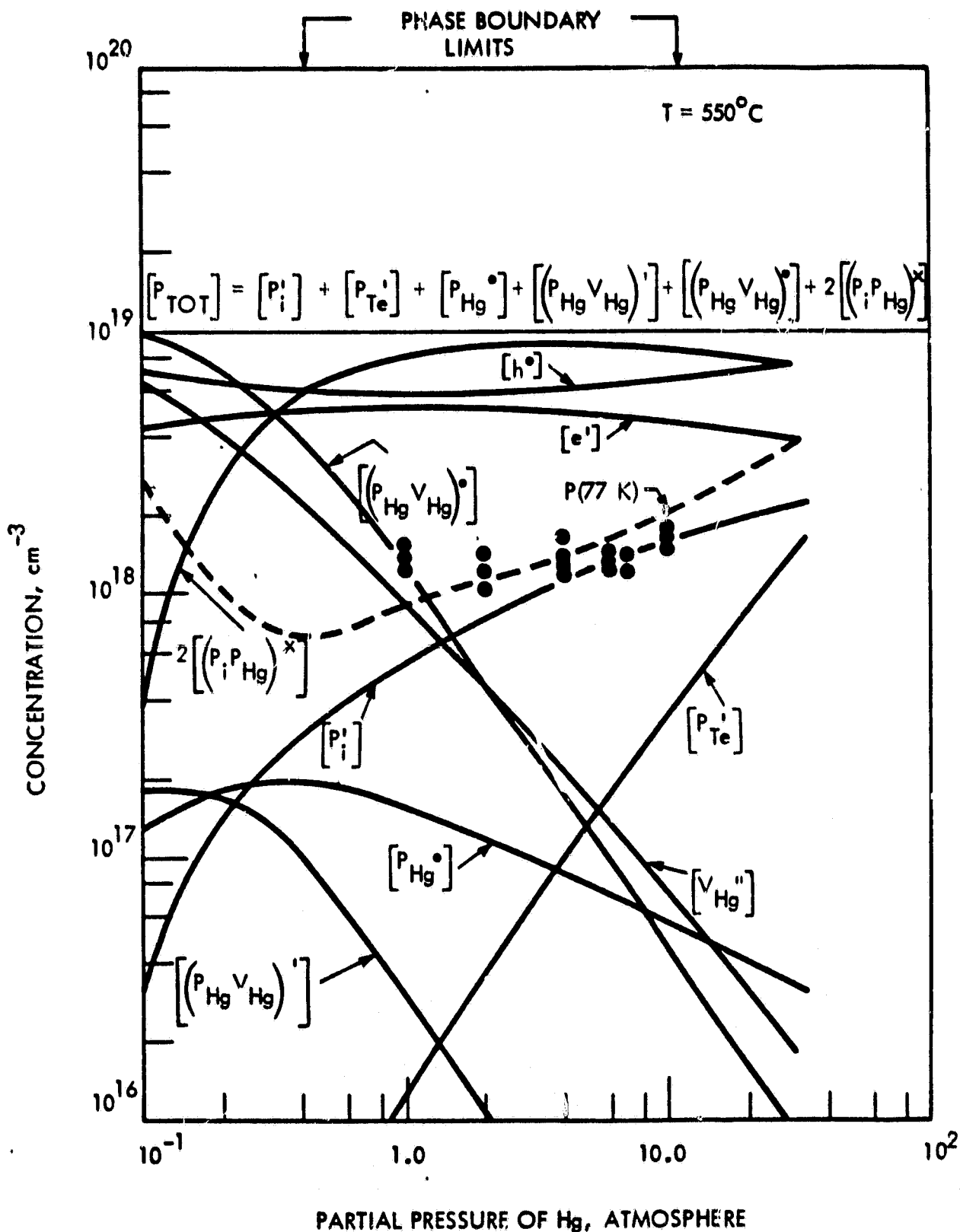


Figure 9. Calculated Concentrations of $[e']$, $[h^\bullet]$, $[V_{\text{Hg}}^{\text{II}}]$, $[P_i']$, $[P_{\text{Te}}']$, $[P_{\text{Hg}}^\bullet]$, $[(P_{\text{Hg}}V_{\text{Hg}})^\bullet]$, $[(P_{\text{Hg}}V_{\text{Hg}})']$ and $[(P_iP_{\text{Hg}})^\times]$ as a Function of the Partial Pressure of Hg at 550°C ; the expected hole concentration in the crystals cooled to 77 K is indicated as a dashed line along with the experimental results.

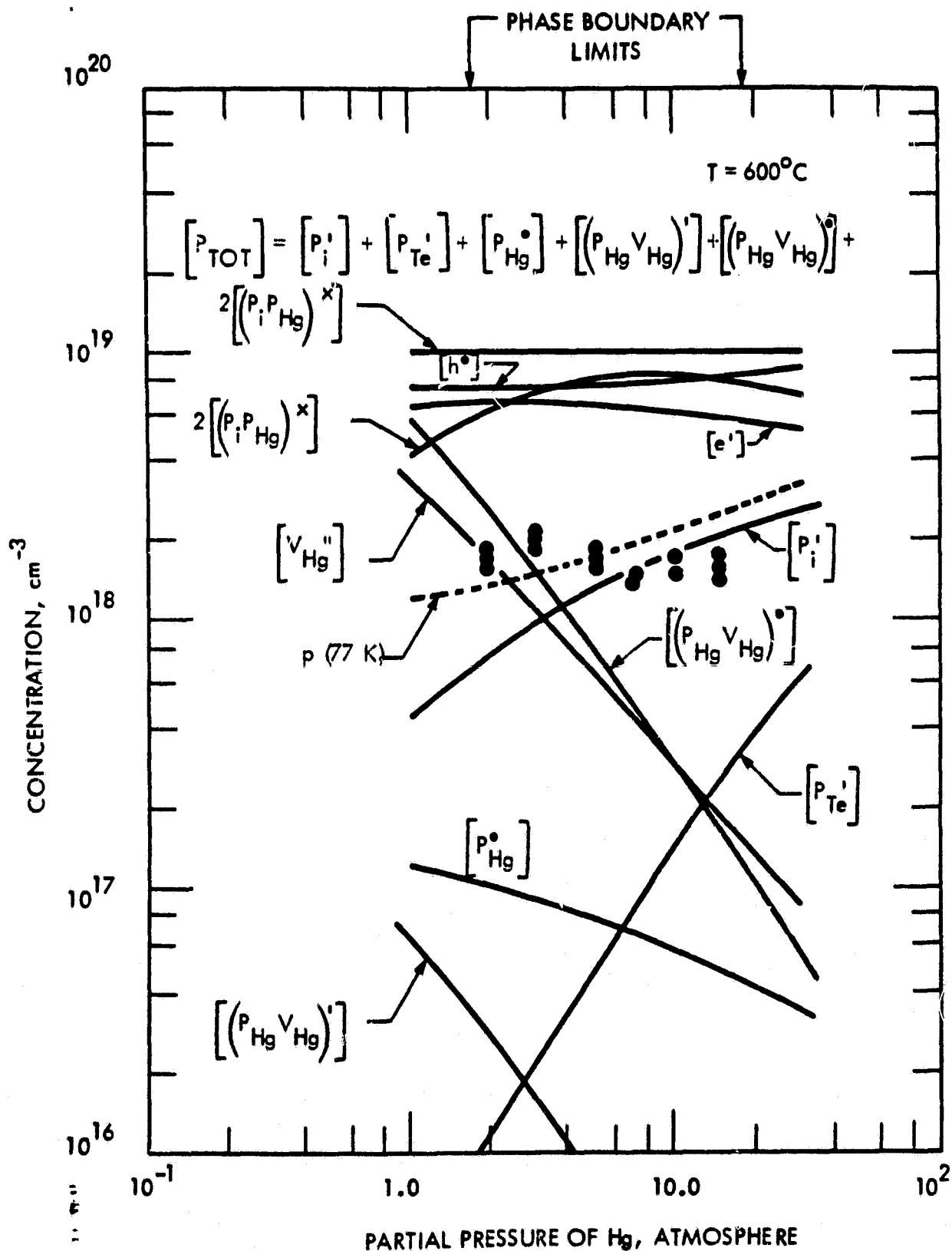


Figure 10. Calculated Concentrations of $[e']$, $[h^{\bullet}]$, $[V_{\text{Hg}}'']$, $[P_i']$, $[P_{\text{Te}}']$, $[P_{\text{Hg}}^{\bullet}]$, $[(P_{\text{Hg}}V_{\text{Hg}})^{\bullet}]$, $[(P_{\text{Hg}}V_{\text{Hg}})']$ and $[(P_iP_{\text{Hg}})^x]$ as a Function of the Partial Pressure of Hg at 600°C ; the expected hole concentration in the crystals cooled to 77 K is indicated as a dashed line along with the experimental results.

defect isotherms show the concentration of the electrons, holes and the various phosphorus defect species as a function of the partial pressure of Hg. The calculated hole concentrations expected at 77 K along with the experimental values are also indicated in the figures. It is apparent from the defect isotherms that the crystal is essentially intrinsic at the annealing temperature except at Hg pressures close to Hg saturated or Te saturated conditions indicated by the phase boundary limits in the figures. It should also be noted that most of the phosphorus is present as neutral $(P_iPHg)^x$ almost throughout the existence region of the crystal except at Hg pressures close to the Hg saturation and Te saturation conditions where the electrically active phosphorus species become significant in concentration. The calculated hole concentrations at 77 K are extremely sensitive to the value of the total phosphorus concentration in the crystals at the lowest Hg pressures where the crystals are very closely compensated (Figures 7 through 10) and the hole concentration in the cooled crystals is much smaller than the concentration of the electrically active native as well as phosphorus defects.

The mass action constants for the incorporation of the various phosphorus species resulting from the present work are listed in Table III.

Table III. VALUES OF THE PARAMETERS FOR THE EQUILIBRIUM CONSTANTS

$K = K_0 \exp (-H/KT)$ defined in Table I.

EQUILIBRIUM CONSTANT	K_0 (SITE FR., ATM)	H (EV)	SOURCE
1 K_i	5.77×10^{-4}	0.57	Ref. 4
2 $K'' V_{Hg}$	7.9×10^2	2.24	
3 $K_{P_{Hg}}$	1.36×10^{-2}	-0.360	This Work
4 $K_{P_{Te}}$	1.81×10^{-26}	-1.73	
5 $K_{P(P_{iP_{Hg}}) \times}$	1.76×10^4	-0.252	
6 $K_{P(P_{Hg} V_{Hg})}$	17	-0.36	
7 $K(P_{Hg} V_{Hg})$	2.1×10^{28}	4.29	

C.2. Coulombic Values for the Pairing Constants $K_P(P_i P_{Hg})^X$ and $K_P(P_{Hg} V_{Hg})'$

The pairing constant is given by $K_P^0 = K_P \exp (-H_P/KT)$. The Coulombic value for the enthalpy of pairing is given by

$$H_P = \frac{-Z_1 Z_2 q^2}{\epsilon r} \quad [\text{ref. 13}] \quad (7)$$

Where Z_1 and Z_2 refer to the charges of the two species comprising the pair, and r to the distance between them.

Noting that $r_{P_i - P_{Hg}} = 3.23 \text{ \AA}$ and

$$r_{P_{Hg} - V_{Hg}} = 4.5 \text{ \AA}$$

$$H_{P(P_i P_{Hg})^X} = -0.252 \text{ eV}$$

$$\text{and } H_{P(P_{Hg} V_{Hg})'} = -0.36 \text{ eV}$$

while analyzing the Hall effect data, the constants $K_P(P_i P_{Hg})^X$ and $K_P(P_{Hg} V_{Hg})'$ were chosen such that their temperature dependences were close to the above coulombic values (Table III). The preexponent or the entropy term (K_P^0) is expected to be ~ 4.0 [13]. However, as can be noted from Table III, the preexponents needed to explain the experimental results are much larger than 4 particularly for $K_P(P_i P_{Hg})^X$.

C.3. Analysis of the Hole Mobility

It is known from the results of previous work [4] that ionized impurity scattering contributes to the mobility of holes at 77 K in $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$. Similar to the procedures used previously, [4] we have undertaken to calculate the hole mobility due to ionized impurity scattering which is then used to calculate the overall mobility by reciprocally combining the mobility due to lattice scattering with the mobility due to ionized impurity scattering. The number of ionized impurity scattering centers obtained at different Hg pressures for different temperatures of anneal as calculated from the defect model is shown in Figures 7 through 10.

For a non-degenerate semiconductor with parabolic bands the mobility due to ionized impurity scattering as given by the Brooks Herring Expression is

$$\mu_I = 2^{7/2} \pi^{-3/2} (kT)^{3/2} (\epsilon_0 \epsilon_s)^2 (m_0)^{-1/2} (m^*/m_0)^{-1/2} N_I^{-1} \left\{ \log_e(1+b) - \frac{b}{1+b} \right\}^{-1}$$

$$\text{where } b = \frac{24 m_0 (m^*/m_0) k^2 (\epsilon_0 \epsilon_s)}{e^2 h^2 p'} \quad (8)$$

$$\text{and } p' = p + (N_A - N_D - p) (p + N_D)/N_A \quad (9)$$

In the above expressions, k is the Boltzmann's constant, T is the temperature, ϵ_0 is the free space permittivity, ϵ_s is the static dielectric constant, m_0 is the free electron mass, (m^*/m_0) is the effective mass ratio for holes, N_I is the number of scattering centers given by the total number of ionized donors and acceptors, e is the electronic charge and h is the planck's constant.

with $T = 77$ K, (m^*/m_0) (holes) $= 0.7$ [16] $\epsilon_s = 17.5$ [16] the expression for the mobility due to ionized impurity scattering becomes

$$\mu_I = 10^{21} N_I^{-1} \left\{ \log_e (1+b) - \frac{b}{1+b} \right\}^{-1} \quad (10)$$

$$\text{and } b = 1.05 \times 10^{19} / p' \quad (11)$$

$$\text{for undoped crystals } N_I = 4 [V_{Hg}^{''}] \quad (12)$$

for phosphorus doped crystals

$$N_I = [P_i'] + [P_{Te}'] + [P_{Hg}^{\bullet}] + [(P_{Hg} V_{Hg})'] + [(P_{Hg} V_{Hg})^{\bullet}] + 4 [V_{Hg}^{''}] \quad (13)$$

The multiplying factor of 4 for the species $V_{Hg}^{''}$ arises in expressions 12 and 13 for N_I since these centers are doubly charged ($z=2$) and hence four times as strong in scattering as the singly charged centers. Since the defects are completely ionized at 77 K in both the undoped crystals and the phosphorus doped crystals (Figure 1)

$$p' = p(77 \text{ K}) = 2 [V_{Hg}^{''}] \text{ (anneal temperature) for the undoped crystals}$$

$$\text{and } p' = p(77 \text{ K}) = [P_i'] + [P_{Te}'] + [(P_{Hg} V_{Hg})'] + 2 [V_{Hg}^{''}] - [P_{Hg}^{\bullet}] - [(P_{Hg} V_{Hg})^{\bullet}]$$

for the phosphorus doped crystals.

From the values of p (77 K) for the phosphorus doped crystals and the concentrations of all the defects as a function of the partial pressure of Hg as given by the defect isotherms of Figures 7 through 10, the hole mobility due to ionized impurity scattering was calculated by reciprocally adding the lattice mobility and the mobility due to ionized impurity scattering. From the results on undoped crystals, the hole mobility due to lattice scattering was assumed to be $\sim 700 \text{ cm}^2/\text{vsec}$ at 77 K for $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ [4]. The results are shown in Figures 11 through 14 where calculated hole mobility is shown as a function of the partial pressure of Hg for various temperatures of anneal; the experimentally measured hole mobilities are also shown in the figures. From the figures, it is apparent that the trend in the variation of the calculated hole mobility as a function of the partial pressure of Hg agrees with the experimental results for the phosphorus doped crystals as well as the undoped crystals. The calculated values for the phosphorus doped crystals decrease drastically at the lowest Hg pressures since the total number of charged defects increases rather strongly under Hg deficient conditions (Figures 7 through 10); in addition, the expected hole concentrations at these low Hg pressures are much lower than for the undoped crystals particularly at 450°C and 500°C. The reduced screening (due to the low hole concentrations) of the coulombic field of the ionized scattering centers along with the rather high concentration of charged defects explains the drastic decrease of the calculated hole mobility at the lowest Hg pressures. However, the experimental hole mobilities are larger than the calculated values at the lowest Hg pressures. The discrepancy can be qualitatively explained by assuming that a fraction of the charged phosphorus defects precipitates during cooling. More sophisticated calculations are needed to explain the results better.

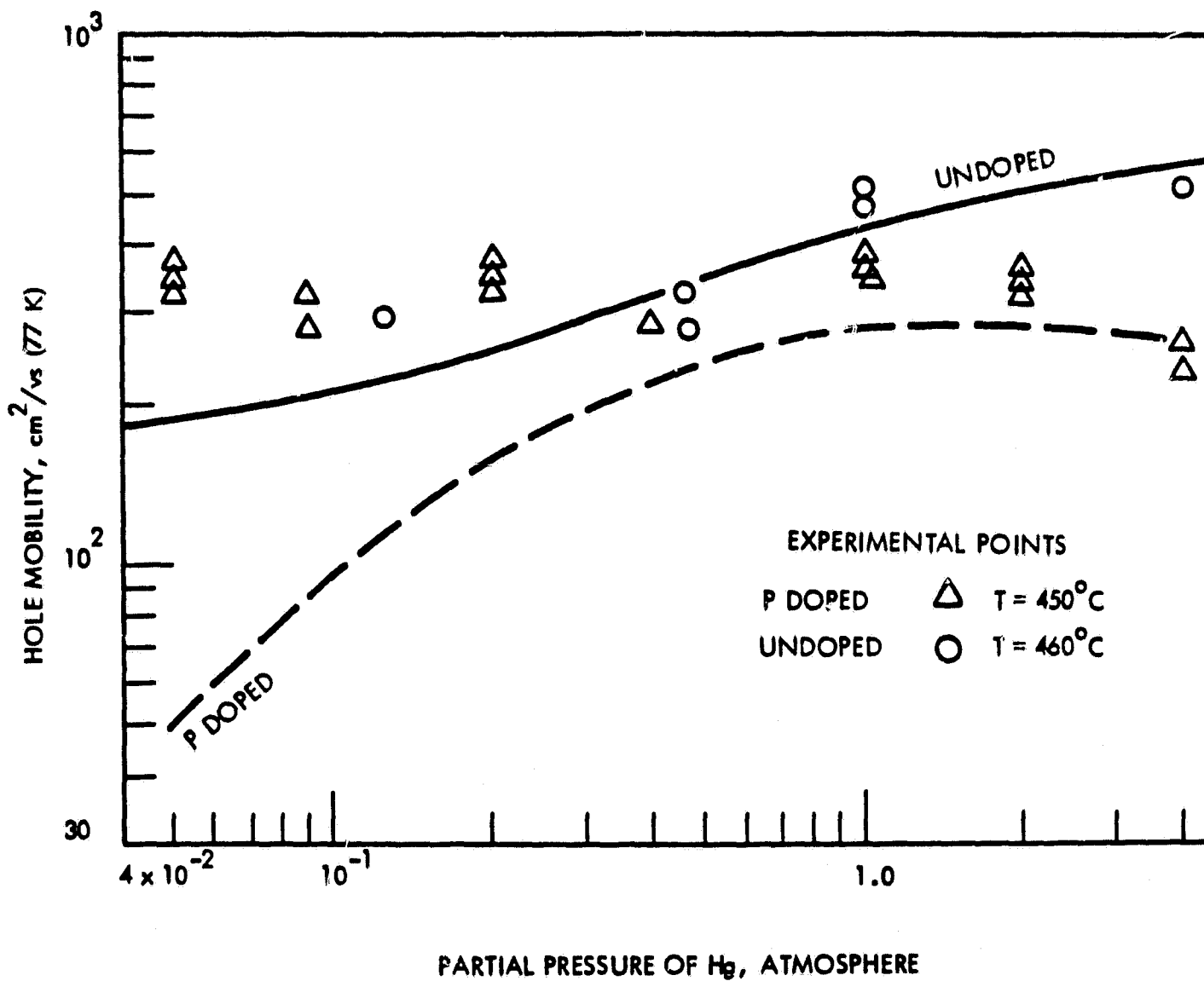


Figure 11. Calculated Hole Mobility at 77 K as a Function of the Partial Pressure of Hg for Phosphorus Doped (10^{19}cm^{-3}) $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ Samples Quenched to Room Temperature After Equilibration at 450°C ; calculation for the undoped crystals after equilibration at 460°C is also shown along with the experimental values for both the doped and the undoped crystals.

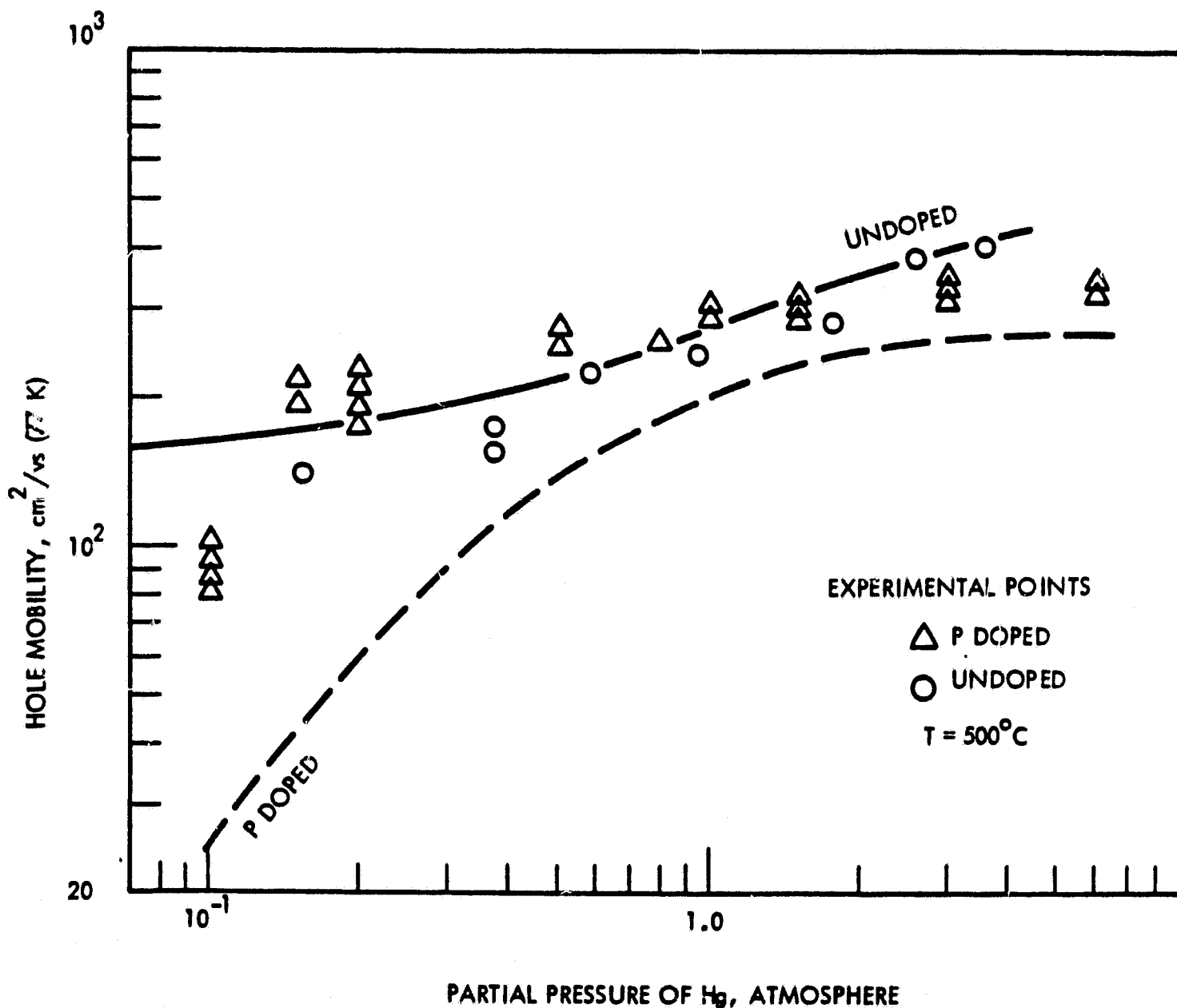


Figure 12. Calculated Hole Mobility at 77 K as a Function of the Partial Pressure of Hg for Phosphorus Doped (10^{19}cm^{-3}) and Undoped $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ Crystals Quenched to Room Temperature After Equilibration at 500°C ; experimental values are also shown for comparison.

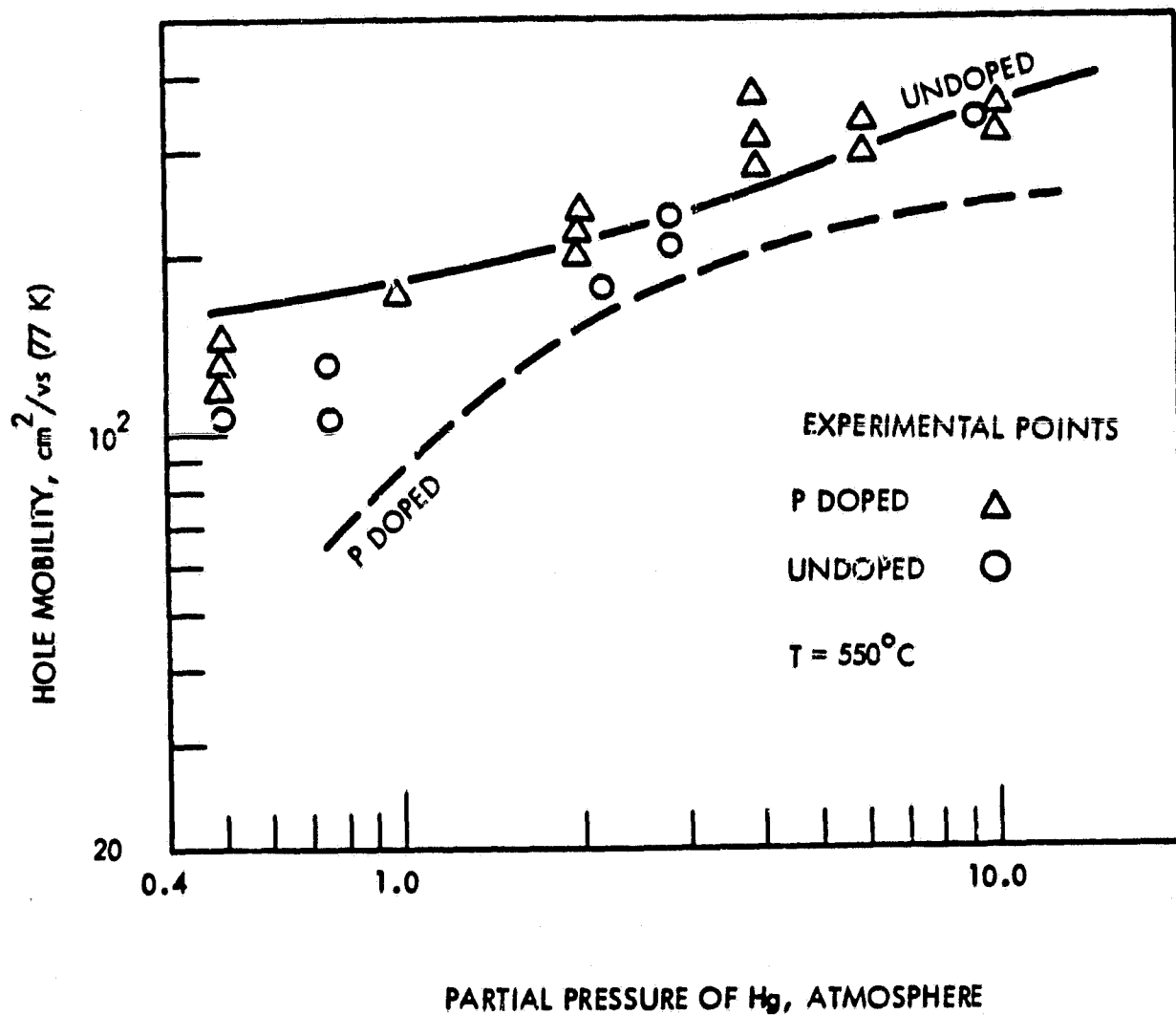


Figure 13. Calculated Hole Mobility at 77 K as a Function of the Partial Pressure of Hg for Phosphorus Doped (10^{19}cm^{-3}) and Undoped $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ Crystals Quenched to Room Temperature After Equilibration at 550°C; experimental values are also shown for comparison.

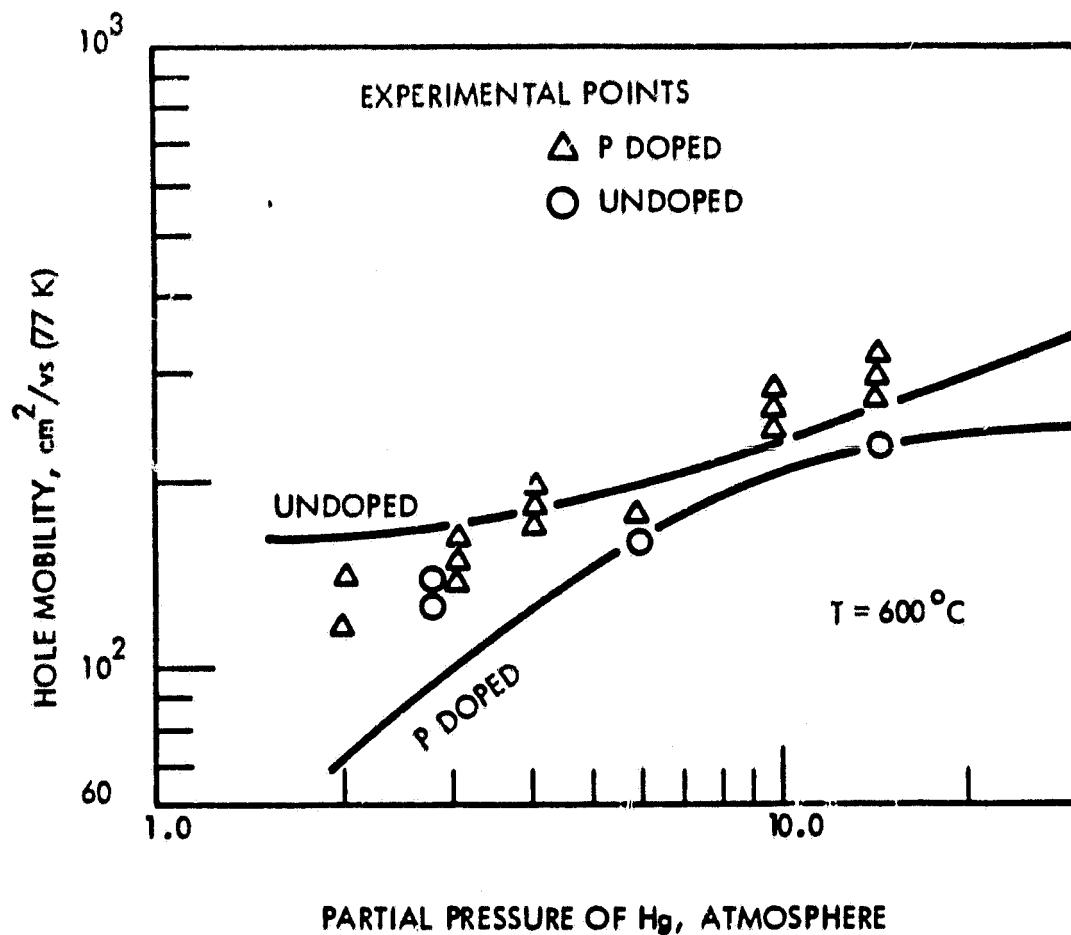


Figure 14. Calculated Hole Mobility at 77 K as a Function of the Partial Pressure of Hg for Phosphorus Doped (10^{19}cm^{-3}) and Undoped $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ Crystals Quenched to Room Temperature After Equilibration at 600°C ; experimental values are also shown for comparison.

D. Comparison with the Defect State in Phosphorus Doped CdTe

The defect model established in the present work for phosphorus doped $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ is very similar to the one established for phosphorus doped CdTe [1]. In both $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ and CdTe, phosphorus occupies interstitial and Te lattices acting as an acceptor while it acts as a donor occupying metal lattice sites. The difference however is that isolated phosphorus donor species are found to be triply ionized in CdTe [1] whereas in $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ it is found to be present only in the singly ionized state. Considering that the band gap of $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ is only 0.1 eV compared with the value of 1.6 eV for CdTe it is understandable that the energy levels of the doubly and triply ionized states of the phosphorus donor species could lie below the valance band edge in $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ whereas they lie within the gap in CdTe. In both $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ and CdTe a large concentration of phosphorus is found to be present as neutral pairs $(\text{P}_{\text{Hg}}\text{P}_{\text{i}})^{\times}$ and $(\text{P}_{\text{Cd}}\text{P}_{\text{i}})^{\times}$ respectively. At low Hg pressures a considerable fraction of phosphorus is found to be present as $(\text{P}_{\text{Hg}}\text{V}_{\text{Hg}})'$ and $(\text{P}_{\text{Hg}}\text{V}_{\text{Hg}})'$ in $(\text{Hg}_{0.8}\text{Cd}_{0.2})\text{Te}$ whereas similar species $(\text{P}_{\text{Cd}}\text{V}_{\text{Cd}})'$ and $(\text{P}_{\text{Cd}}\text{V}_{\text{Cd}})'$ have not been reported for CdTe [1].

V. SUMMARY

Phosphorus doped $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ single crystals were annealed at 450°C to 600°C in different partial pressures of Hg. Hall effect measurements were made on crystals quenched to room temperature after the anneals. The results are explained on the basis of a defect model in which phosphorus behaves amphoterically acting as a single acceptor in interstitial sites at high Hg pressures and occupying Te lattice sites ($\text{P}_{\text{i}}^{\text{i}}$ and $\text{P}_{\text{Te}}^{\text{i}}$) and as a single donor occupying Hg lattice sites ($\text{P}_{\text{Hg}}^{\text{o}}$). Most of the phosphorous appears to be present as neutral pairs $(\text{P}_{\text{i}}\text{P}_{\text{Hg}})^{\times}$ at moderate Hg pressures while a considerable fraction also exists paired with the native acceptor defects $(\text{P}_{\text{Hg}}\text{V}_{\text{Hg}})^{\cdot}$ and $(\text{P}_{\text{Hg}}\text{V}_{\text{Hg}})^{\circ}$ at low Hg pressures. Equilibrium constants for the incorporation of the various phosphorous species have been evaluated.

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IV. SUMMARY

The work performed on this program so far has resulted in the establishment of the defect models for undoped $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ and undoped $\text{Hg}_{0.6}\text{Cd}_{0.4}\text{Te}$. The mechanisms of incorporation of copper, indium, iodine and phosphorus in $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ have also been established. In the next semiannual report, the details of incorporation of gold in $\text{Hg}_{0.8}\text{Cd}_{0.2}\text{Te}$ will be discussed.